

Atty. Docket #: PH 99012 G1

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

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**INTERNATIONAL APPL. NO.:** PCT/EP00/01102 :**INTERNATIONAL FILING DATE:** -02/01/2000- :**APPLICANT:** DAVID ALAN ROBERTS ET AL :**SERIAL NO:** (To be assigned) : **ART UNIT:****FILED:** -HEREWITH- : **EXAMINER:****FOR:** "WEED CONTROL" :  
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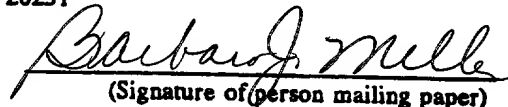
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Commissioner for Patents  
Box PCT  
Washington, D.C. 20231

**"Express Mail" No.:** EK954093795**Date:** -JULY 30, 2001-

I hereby certify that this paper, along with any other paper or fee referred to in this paper as being transmitted herewith, is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10, postage prepaid, on the date indicated above, addressed to Box PCT, Commissioner for Patents, Washington, D.C. 20231

-Barbara J. Miller-  
(Typed or printed name of mailing paper or fee)

  
(Signature of person mailing paper)

**TRANSMITTAL OF APPLICATION PAPERS  
TO U.S. DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. §371.  
(37 CFR 1.494 OR 1.495)**

This Transmittal Letter is based upon PTO Form 1390 (as revised in May, 1993).

The above-identified applicant(s) (jointly with their assignee) have filed an International Application under the P.C.T. and hereby submit(s) to the United States Designated/Elected Office (DO/EO/US) the following items and other information.

JC18 Rec'd PCT/PTO 30 JUL 2001

1. ☒ This is a FIRST submission of items concerning a filing under 35 U.S.C. §371.
2. ☐ This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. §371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay.
4. ☒ A proper Demand for International Preliminary Examination (IPE) was made to the appropriate Authority (IPEA) within the time period required.
5. ☒ A copy of the International Application as filed (35 U.S.C. §371(c)(2)) --  
a. ☒ is transmitted herewith (required when not transmitted by International Bureau). -IN ENGLISH-. See WIPO Publication  
b. ☐ has been transmitted by the International Bureau. WO 00/45637  
c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A (verified) translation of the International Application into the English language is enclosed. (See No. 5. above)
7. ☐ Amendments to the (specification and) claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))  
a. ☐ are transmitted herewith (required if not transmitted by the International Bureau).  
b. ☐ have been transmitted by the International Bureau.  
c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.  
d. ☐ have not been made and will not be made.  
e. ☐ will be submitted with the appropriate surcharge.
8. ☐ A translation of the amendments to the claims (and/or the specification) under PCT Article 19 (35 U.S.C. §371(c)(3)) is enclosed or will be submitted with the appropriate surcharge.

International Application No. PCT/EP00/01102

9. ☒ An oath or declaration/power of attorney of the inventor(s) (35 U.S.C. §371(c)[4]) will follow.  
[ ] and is attached to the translation of (or a copy of) the International Application.  
[ ] and is attached to the substitute specification.

10. ☒ A translation of at least the Annexes to the IPE Report under PCT Article 36 (35 U.S.C. §371(c)[5]) is enclosed. (PCT/IPEA/416 & 409)

Items 11. to 16. below concern other document(s) or information included:

11. [ ] An Information Disclosure Statement under 37 CFR 1.97 and 1.98 is enclosed.  
12. ☒ An Assignment for recording and a separate cover sheet in compliance with 37 CFR 3.28 and 3.31 will follow.  
13. ☒ A FIRST preliminary amendment is enclosed.  
A SECOND or SUBSEQUENT preliminary amendment is enclosed.  
14. [ ] A substitute specification (including claims, abstract, drawing) is enclosed.  
15. [ ] A change of power of attorney and/or address letter is enclosed.  
16. ☒ Other items of information:

- ☒ This application is being filed pursuant to 37 CFR 1.494(c) or 1.495(c), and any missing parts will be filed before expiration of--

[ ] 22 months from the priority date under 37 CFR 1.494(c), or

☒ 32 months from the priority date under 37 CFR 1.495(c).

- ☒ The undersigned attorney is authorized by the International applicant and by the inventors to enter the National Phase pursuant to 37 CFR 1.494(c) or 1.495(c).

The following additional information relates to the International Application:

International Application No. PCT/EP00/01102

PH 99012 G1

- ☒ Receiving Office: EPO  
☒ IPEA (if filing under 37 CFR 1.495): EPO  
☒ Priority Claim(s) (35 USC §§ 119, 365):  
Great Britain Appln. 9902232.9 filed -February 01, 1999- AND  
9908313.1 filed -April 12, 1999.  
☒ A copy of the International Search Report is

☐ enclosed.

☒ attached to the copy of the International  
Application.

☒ A copy of the Receiving Office Request Form is enclosed.

- [X] Form PCT/IB/304 (1) sheet  
[X] Form PCT/IB/308 (2) sheets  
[X] Form PCT/IB/332 (1) sheet

The fee calculation is set forth on the next page of this Transmittal Letter.

International Application No. PCT/EP00/01102

### FEE CALCULATION SHEET

☒ A check in payment of the filing fee, calculated as follows, is attached (37 CFR 1.492).

Basic Fee..... \$ 860.00

Total Number of claims in  
excess of (20) times \$18..... -0-

Number of independent claims  
in excess of (3) times \$78..... -0-

Fee for multiple dependent  
claims \$260..... -0-

TOTAL FILING FEE... \$ 860.00

Kindly send us the official filing receipt.

The Commissioner is hereby authorized to charge any additional fees which may be required or to credit any overpayment to Deposit Account No. 03-2775. This is a "general authorization" under 37 CFR 1.25(b), except that no automatic debit of the issue fee upon allowance is authorized. An additional copy of this page is attached.

Respectfully submitted,

By



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WEM/bjm (5500\*99)  
Enclosures

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PH 99012 G1  
5500\*99

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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**DAVID ALAN ROBERTS, ET AL.** : **PCT/EP00/01102**  
**SERIAL NO.:** **To Be Assigned** : **ART UNIT: TBA**  
**FILED:** **Herewith** : **EXAMINER: TBA**  
**FOR:** **WEED CONTROL** :  
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Commissioner for Patents  
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"Express Mail" No.: EK954093795

Date: July 30, 2001

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Barbara J. Miller



(Typed or printed name of person mailing paper or fee)

(Signature of person mailing paper or fee)

**PRELIMINARY AMENDMENT**

Sir:

Prior to the determination of the filing fee and any action on the merits of the accompanying new patent application, kindly amend the application as follows:

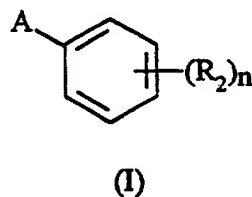
**In the Claims:**

Please cancel claims 12 and 13 without prejudice or disclaimer.

Please amend claims 3-6, 8-10 and 12-13 as follows. A clean copy of the amended claims is provided as Appendix A.

3. (Amended) A method according to claim 1 [or 2] in which the locus is a crop-growing locus.

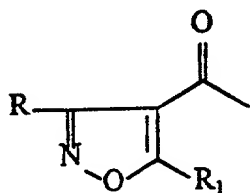
4. (Amended) A method according to claim 1 [, 2 or 3] in which the surface layer of the medium is from the surface to a depth of 10 cm.
5. (Amended) A method according to [any one of the preceding claims] claim 1 which comprises applying to the locus sequential low doses of isoxazole herbicide.
6. (Amended) A method according to [any one of claims 1 to 4] claim 1 which comprises treating the locus with a delayed release composition comprising the isoxazole herbicide.
8. (Amended) A method according to claim 6 [or 7] in which an encapsulated isoxazole is used, comprising an isoxazole derivative encapsulated with a solid film comprising an inert material itself having no substantial herbicidal activity.
9. (Amended) A method according to claim 8 in which [granules of an] the isoxazole derivative [derivate] is in the form of granules of from 0.1 to 50  $\mu\text{m}$  in size [are used].
10. (Amended) A method according to [any one of the preceding claims] claim 1 in which the isoxazole derivative is of general formula I [as hereinbefore defined]:



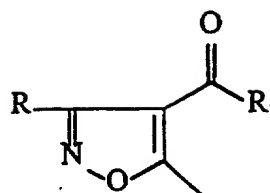
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wherein:

A represents a group (A-1) or (A-2):



(A-1)



(A-2)

wherein:

R represents a hydrogen atom or a halogen atom; a straight- or branched-chain alkyl or alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; a cycloalkyl group containing from 3 to 6 carbon atoms optionally substituted by one or more groups R<sup>5</sup>, one or more halogen atoms or a group -CO<sub>2</sub>R<sup>3</sup>; or a group selected from -CO<sub>2</sub>R<sup>3</sup>, -COR<sup>5</sup>, cyano, nitro, -CONR<sup>3</sup>R<sup>4</sup> and -S(O)<sub>k</sub>R<sup>13</sup>.

R<sup>1</sup> represents a straight- or branched-chain alkyl, alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms, or a cycloalkyl group containing from three to six carbon atoms optionally substituted by one or more groups R<sup>5</sup> or one or more halogen atoms;

R<sup>2</sup> represents a halogen atom; a straight- or branched-chain alkyl, alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; a



straight- or branched-chain alkyl group containing up to six carbon atoms which is substituted by one or more groups -OR<sup>5</sup>; or a group selected from nitro, cyano, -CO<sub>2</sub>R<sup>5</sup>, -S(O)<sub>p</sub>R<sup>6</sup>, -O(CH<sub>2</sub>)<sub>m</sub>OR<sup>5</sup>, -COR<sup>5</sup>, -NR<sup>11</sup>R<sup>12</sup>, -N(R<sup>8</sup>)SO<sub>2</sub>R<sup>7</sup>, -N(R<sup>8</sup>)CO<sub>2</sub>R<sup>7</sup>, -OR<sup>5</sup>, -OSO<sub>2</sub>R<sup>7</sup>, -SO<sub>2</sub>NR<sup>3</sup>R<sup>4</sup>, -CONR<sup>3</sup>R<sup>4</sup>, -CSNR<sup>3</sup>R<sup>4</sup>, -(CR<sup>9</sup>R<sup>10</sup>)<sub>i</sub>-S(O)<sub>q</sub>R<sup>7</sup> and -SF<sub>5</sub>; or two groups R<sup>2</sup>, on adjacent carbon atoms of the phenyl ring may, together with the carbon atoms to which they are attached, form a 5 to 7 membered saturated or unsaturated heterocyclic ring containing up to three ring heteroatoms selected from nitrogen, oxygen and sulfur, which ring is optionally substituted by one or more groups selected from halogen, nitro, -S(O)<sub>p</sub>R<sup>13</sup>, C<sub>1-4</sub> alkyl, C<sub>1-4</sub> alkoxy, C<sub>1-4</sub> haloalkyl, C<sub>1-4</sub> haloalkoxy, =O (or a 5- or 6-membered cyclic acetal thereof), and =NO-R<sup>3</sup>, it being understood that a sulphur atom, where present in the ring, may be in the form of a group -SO- or -SO<sub>2</sub>;

n represents an integer from one to five; when n is greater than one the groups R<sup>2</sup> may be the same or different;

R<sup>3</sup> and R<sup>4</sup> each independently represent a hydrogen atom, or a straight- or branched-chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms;

R<sup>5</sup> represents a straight- or branched-chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; or a straight- or branched-chain alkenyl or alkynyl group containing from two to six carbon atoms which is optionally substituted by one or more halogen atoms;

R<sup>6</sup> and R<sup>7</sup>, which may be the same or different, each represent R<sup>5</sup> or phenyl optionally substituted by from one to five groups which may be the same or different selected from a halogen atom, a straight- or branched-chain alkyl group containing up to six carbon atoms which is

optionally substituted by one or more halogen atoms, nitro, cyano,  $-\text{CO}_2\text{R}^5$ ,  $-\text{S}(\text{O})_p\text{R}^{13}$ , -

$\text{NR}^{11}\text{NR}^{12}$ ,  $-\text{OR}^5$ , and  $-\text{CONR}^3\text{R}^4$ ;

$\text{R}^8$ ,  $\text{R}^9$  and  $\text{R}^{10}$  each represent a hydrogen atom or  $\text{R}^6$ ;

$\text{R}^{11}$  and  $\text{R}^{12}$  each represent hydrogen or  $\text{R}^5$ ;

$\text{R}^{13}$  represents a straight- or branched-chain alkyl group containing up to six carbon atoms

which is optionally substituted by one or more halogen atoms;

k, p and q independently represent the values zero, one or two;

m represents one, two or three;

t represents an integer from one to four; when t is greater than one, the groups  $\text{R}^9$  and  $\text{R}^{10}$

may be the same or different;

or an agriculturally acceptable salt or metal complex thereof.

Please add new claims 14 to 17 as shown below.

-- 14. A method according to claim 1, wherein the isoxazole herbicide is selected from the group consisting of:

5-cyclopropyl-4-[2-chloro-3-ethoxy-4-(ethylsulphonyl)benzoyl]isoxazole;

4-(4-chloro-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;

5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole;

4-(4-bromo-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;

5-cyclopropyl-4-[4-fluoro-3-methoxy-2-(methylsulphonyl)benzoyl]isoxazole;

4-(4-bromo-2-methylsulphonylmethylbenzoyl)-5-cyclopropylisoxazole;

ethyl 5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole-3-

carboxylate; and

5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)-3-methylthio-isoxazole.

15. A delayed release composition according to claim 11, wherein the isoxazole herbicide is selected from the group consisting of:

5-cyclopropyl-4-[2-chloro-3-ethoxy-4-(ethylsulphonyl)benzoyl]isoxazole;

4-(4-chloro-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;

5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole;

4-(4-bromo-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;

5-cyclopropyl-4-[4-fluoro-3-methoxy-2-(methylsulphonyl)benzoyl]isoxazole;

4-(4-bromo-2-methylsulphonylmethylbenzoyl)-5-cyclopropylisoxazole;

ethyl 5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole-3-

carboxylate; and

5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)-3-methylthio-isoxazole.

16. A method according to claim 1, wherein the isoxazole herbicide is isoxaflutole.

17. A delayed release composition according to claim 11, wherein the isoxazole herbicide is isoxaflutole.- -

**REMARKS**

Claims 3-6 and 8-10 have been amended. Claims 12 and 13 have been canceled without prejudice or disclaimer. Each of the dependent claims, as amended, now depends on only one preceding claim. Therefore no additional fee is required for multiple dependency.

New claims 14 to 17 have been added. It is respectfully submitted that no new matter has been introduced into the present application by any of the amendments or by the addition of the new claims.

Prompt, favorable action is solicited.

Respectfully submitted,

CONNOLLY BOVE LODGE & HUTZ LLP

By: 

William E. McShane  
Registration No. 32,707  
P. O. Box 2207  
Wilmington, Delaware 19899  
(302) 888-6248  
Attorney for Applicants

WEM:bjm

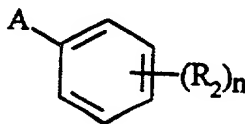
Enclosure: Appendix A - clean copy of amended and new claims

::ODMA\MHODMA\CB;155415,1

APPENDIX A

Clean Copy of Amended and New Claims

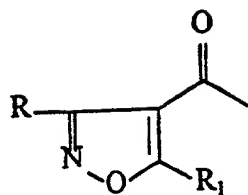
3. A method according to claim 1 in which the locus is a crop-growing locus.
4. A method according to claim 1 in which the surface layer of the medium is from the surface to a depth of 10 cm.
5. A method according to claim 1 which comprises applying to the locus sequential low doses of isoxazole herbicide.
6. A method according to claim 1 which comprises treating the locus with a delayed release composition comprising the isoxazole herbicide.
8. A method according to claim 6 in which an encapsulated isoxazole is used, comprising an isoxazole derivative encapsulated with a solid film comprising an inert material itself having no substantial herbicidal activity.
9. A method according to claim 8 in which the isoxazole derivative is in the form of granules of from 0.1 to 50  $\mu\text{m}$  in size.
10. A method according to claim 1 in which the isoxazole derivative is of general formula I :



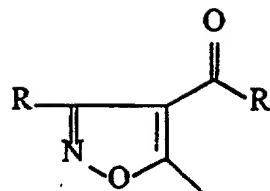
(I)

wherein:

A represents a group (A-1) or (A-2):



(A-1)



(A-2)

wherein:

R represents a hydrogen atom or a halogen atom; a straight- or branched-chain alkyl or alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; a cycloalkyl group containing from 3 to 6 carbon atoms optionally substituted by one or more groups  $R^5$ , one or more halogen atoms or a group  $-CO_2R^3$ ; or a group selected from  $-CO_2R^3$ ,  $-COR^5$ , cyano, nitro,  $-CONR^3R^4$  and  $-S(O)_kR^{13}$ ;

$R^1$  represents a straight- or branched-chain alkyl, alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms, or a cycloalkyl group containing from three to six carbon atoms optionally substituted by one or more groups  $R^5$  or one or more halogen atoms;

$R^2$  represents a halogen atom; a straight- or branched-chain alkyl, alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; a

straight- or branched-chain alkyl group containing up to six carbon atoms which is substituted by one or more groups  $-OR^5$ ; or a group selected from nitro, cyano,  $-CO_2R^5$ ,  $-S(O)_pR^6$ ,  $-O(CH_2)_mOR^5$ ,  $-COR^5$ ,  $-NR^{11}R^{12}$ ,  $-N(R^8)SO_2R^7$ ,  $-N(R^8)CO_2R^7$ ,  $-OR^5$ ,  $-OSO_2R^7$ ,  $-SO_2NR^3R^4$ ,  $-CONR^3R^4$ ,  $-CSNR^3R^4$ ,  $-(CR^9R^{10})_t-S(O)_qR^7$  and  $-SF_5$ ; or two groups  $R^2$ , on adjacent carbon atoms of the phenyl ring may, together with the carbon atoms to which they are attached, form a 5 to 7 membered saturated or unsaturated heterocyclic ring containing up to three ring heteroatoms selected from nitrogen, oxygen and sulfur, which ring is optionally substituted by one or more groups selected from halogen, nitro,  $-S(O)_pR^{13}$ ,  $C_{1-4}$  alkyl,  $C_{1-4}$  alkoxy,  $C_{1-4}$  haloalkyl,  $C_{1-4}$  haloalkoxy,  $=O$  (or a 5- or 6-membered cyclic acetal thereof), and  $=NO-R^3$ , it being understood that a sulphur atom, where present in the ring, may be in the form of a group  $-SO-$  or  $-SO_2-$ ;

$n$  represents an integer from one to five; when  $n$  is greater than one the groups  $R^2$  may be the same or different;

$R^3$  and  $R^4$  each independently represent a hydrogen atom, or a straight- or branched-chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms;

$R^5$  represents a straight- or branched-chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; or a straight- or branched-chain alkenyl or alkynyl group containing from two to six carbon atoms which is optionally substituted by one or more halogen atoms;

$R^6$  and  $R^7$ , which may be the same or different, each represent  $R^5$  or phenyl optionally substituted by from one to five groups which may be the same or different selected from a halogen atom, a straight- or branched-chain alkyl group containing up to six carbon atoms which is

optionally substituted by one or more halogen atoms, nitro, cyano,  $-\text{CO}_2\text{R}^5$ ,  $-\text{S}(\text{O})_p\text{R}^{13}$ , -

$\text{NR}^{11}\text{NR}^{12}$ ,  $-\text{OR}^5$ , and  $-\text{CONR}^3\text{R}^4$ ;

$\text{R}^8$ ,  $\text{R}^9$  and  $\text{R}^{10}$  each represent a hydrogen atom or  $\text{R}^6$ ;

$\text{R}^{11}$  and  $\text{R}^{12}$  each represent hydrogen or  $\text{R}^5$ ;

$\text{R}^{13}$  represents a straight- or branched-chain alkyl group containing up to six carbon atoms

which is optionally substituted by one or more halogen atoms;

k, p and q independently represent the values zero, one or two;

m represents one, two or three;

t represents an integer from one to four; when t is greater than one, the groups  $\text{R}^9$  and  $\text{R}^{10}$  may be the same or different;

or an agriculturally acceptable salt or metal complex thereof.

14. A method according to claim 1, wherein the isoxazole herbicide is selected from the group consisting of:

5-cyclopropyl-4-[2-chloro-3-ethoxy-4-(ethylsulphonyl)benzoyl]isoxazole;

4-(4-chloro-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;

5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole;

4-(4-bromo-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;

5-cyclopropyl-4-[4-fluoro-3-methoxy-2-(methylsulphonyl)benzoyl]isoxazole;

4-(4-bromo-2-methylsulphonylmethylbenzoyl)-5-cyclopropylisoxazole;

ethyl 5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole-3-carboxylate; and

5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)-3-methylthio-isoxazole.



15. A delayed release composition according to claim 11, wherein the isoxazole herbicide is selected from the group consisting of:

5-cyclopropyl-4-[2-chloro-3-ethoxy-4-(ethylsulphonyl)benzoyl]isoxazole;

4-(4-chloro-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;

5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole;

4-(4-bromo-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;

5-cyclopropyl-4-[4-fluoro-3-methoxy-2-(methylsulphonyl)benzoyl]isoxazole;

4-(4-bromo-2-methylsulphonylmethylbenzoyl)-5-cyclopropylisoxazole;

ethyl 5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole-3-

carboxylate; and

5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)-3-methylthio-isoxazole.

16. A method according to claim 1, wherein the isoxazole herbicide is isoxaflutole.

17. A delayed release composition according to claim 11, wherein the isoxazole herbicide is isoxaflutole.

## WEED CONTROL

5 This invention relates to a method for controlling the growth of weeds by the progressive application to, or release into, the surface layer of a growing medium, of a benzoylisoxazole herbicide, and to compositions for use in the method.

### **Background of the invention**

10 The use of isoxazoles for controlling weeds has been described in European Patent Publication Nos. 0418175, 0487357, 0527036 and 0560482. The herbicidal activity of diketonitriles (DKN) which may be formed from the isoxazoles has also been described in European Patent Publication Nos. 15 0213892, 0496630 and 0496631, and International Publication No. WO 95/25099.

Following the application of isoxazole herbicides they may decompose to diones, in particular diketonitrile (DKN) compounds. This conversion is generally irreversible. The DKN compounds are generally also herbicides. 20 They are generally more water soluble than the isoxazole herbicides and may be subject to movement in the soil profile following rainfall.

It has been found that by modifying the way isoxazole herbicides are applied the ratio of isoxazole to DKN in a growing medium such as soil can be altered in favour of the isoxazole, increasing the ratio of isoxazole to 25 DKN.

It has been found that by maintaining the ratio of isoxazole to DKN, in favour of the isoxazole, in the surface layer of the soil, for example, during the period from application of isoxazole to establishment of a crop, improved control of weeds may be obtained. Furthermore, crop selectivity may be 30 improved and the risk of run off and leaching may be reduced.

An object of the present invention is to provide a method of application and/or composition which reduces the net movement of isoxazole and DKN

through the soil and retains the compounds in the soil surrounding, preferably immediately surrounding, the point of application of the isoxazole.

Another object of the present invention is to provide a method and/or composition which permits the delivery of lower individual dose rates of isoxazole herbicides while maintaining (and sometimes improving) herbicidal efficacy.

A further object of the present invention is to provide a composition, comprising an isoxazole, with improved activity on weed species and/or improved crop selectivity.

The objects of the invention can be achieved in whole or in part by the present invention.

It is known that isoxazoles exert their herbicidal activity in plants by conversion to DKN compounds. It might be expected, therefore, that application of isoxazole in such a way as to accelerate or favour its conversion to DKN would be advantageous. The Applicants have found that the opposite is true.

The present invention provides a method for controlling the growth of weeds at a locus in a solid growing medium which comprises treating the locus with a composition comprising an isoxazole herbicide to provide progressive or sequential delivery or release of isoxazole herbicide into the surface layer of the medium.

The growing medium includes composts but is preferably the soil.

The locus is preferably a crop-growing locus, for example, where a crop is sown and cultivated.

The surface layer is generally from the surface to a depth of 10 cm, preferably to a depth of 5 cm, more preferably to a depth of 3 cm.

According to a feature of the invention the method comprises applying to the locus, for example where a crop is sown and cultivated, sequential low doses of isoxazole herbicides. For example the normal dosage may be divided into two or more, for example 2 to 5, generally equal portions and applied at time-spaced intervals, each application after the first being made, for example 1 to 4 days, preferably 1 day, after the preceding one.

According to a further feature of the present invention the method comprises treating the locus with a delayed release composition comprising the isoxazole herbicide.

The delayed release composition, which constitutes a feature of the invention, may comprise, for example, an encapsulated composition comprising the isoxazole itself or a composition containing it. The delayed release compositions may be prepared by known methods.

The encapsulated product may have a solid outer wall, said wall comprising an inert material, generally having no substantial herbicidal activity.

The encapsulated isoxazole according to the present invention may comprise granules comprising an isoxazole derivative of formula (I), each of these granules being encapsulated with a solid film comprising an inert material itself having no substantial herbicidal activity.

Preferably the inert material is a water-soluble polymeric material, modified by treatment to render it substantially water insoluble.

Soluble materials which may used include:

a copolyester; polyvinylalcohol; polyacrylate; polycarboxylate; gelatine; polysulfonate, for example the polystyryl polysulfones, a protein, a polyethylene oxide; a modified or unmodified starch; a cellulose for example carboxymethyl cellulose; a dextran, maltose, an alkyl-, hydroxyalkyl-, carboxyalkyl-cellulose; a polyvinylether; poly(2,4-diethyl-6-triazoethylen); poly(vinylsulfonic acid), polyanhydride, a low molecular weight urea-formaldehyde resin, a low molecular weight melamine-formaldehyde resin, a polymethacrylate for example poly(alkylcyanoacrylate), poly(isobutylcyanoacrylate), poly(2-hydroxyethylmethacrylate), polyacrylic acid or a homologue thereof; low molar mass amphiphiles; low molar mass polymeric amphiphiles; polylactic acid glutamic acid; dendrimers (hyperbranched polymers); phospholipids for example distearoylphosphatidylcholine, dioleoylphosphatidylethanolamine, dipalmitoylphosphatidylcholine, dipalmitoylphosphatidylglycerol, phosphatidylethanolamine, phosphatidylinositol; lipoprotein, semi-solid poly(orthoester)

polycarboxylates; hydrogels. The materials may be in the form of, for example, solid lipid nano/micro spheres; polyester microspheres, nanocapsules, niosomes, liposomes, polymeric micelles. An oil may be used to facilitate the production of an emulsion with small particle sizes and to inhibit agglomeration.

Preferably the water-soluble material is a copolyester, for example gerol which is a copolymer for example: 1,3-Benzenedicarboxylic acid, 5-sulfo-monosodium salt, polymer with 1,3-benzenedicarboxylic acid, 1,4-benzenedicarboxylic acid, 1,2-ethanediol, 2,2'-[1,2-ethanediylbis(oxy)]bis[ethanol] and 2,2'-oxybis[ethanol]. The water-soluble materials may vary in molecular weight and may include oligomers.

The inert water-soluble polymer is generally precipitated by association (by complexation or mixing) with a material which does not itself solubilise the aforementioned water-soluble polymer. The materials which result in precipitation of the polymer include soluble salts of alkaline earth metals (for example calcium). The association can be modulated by adjusting the pH of the water soluble solution of the polymer which solubilises the ions (of the aforementioned alkaline earth) which effect precipitation of the now insolubilised polymer to encapsulate the particles of the active material. The pH can be adjusted using, for example, acetic acid. Precipitation can also be induced by adjusting the solvent or solvents without the need for association with another material.

The size of the granules of the active material of an isoxazole derivative of formula (I) is generally from 0.1 to 50  $\mu\text{m}$ , preferably from 1 to 20  $\mu\text{m}$ .

The thickness of the coating of the encapsulating material is generally from 0.1 to 50  $\mu\text{m}$ , preferably from 1 to 20  $\mu\text{m}$ .

The granules of encapsulated 4-benzoylisoxazole derivative of formula (I) according to the present invention may be for example, in a powdered state or in a liquid or solid formulation, contained within a support (or a carrier for application).

Compositions of the present invention improve the release of a herbicide to the soil site to which it is applied and movement resulting from

rain water or irrigation is reduced. A marked improvement in the downward transmigration of the active ingredient from the immediate application area (weed seed zone) through the soil profile is provided.

Compositions of the present invention provide a method for controlling the release of a herbicide in a range of soil types and edaphic conditions by modification of the ratios of isoxazole: carrier material.

The use of compositions of the present invention permits more efficient use of herbicide which is retained in the area of soil application, for example the weed seed zone; thus the amount of herbicide applied may be reduced. In addition, the herbicide is maintained in the area of the weed seed zone for a longer period of time than usual.

Localisation of the herbicide in the weed seed zone improves selectivity on the crop species, for example, maize.

Compositions of the present invention provide a method for reducing the release rate of herbicides, such as isoxazoles, in soil thereby slowing the overall rate of degradation. Encapsulation in a matrix carrier increases the stability of the herbicides as it is protected from the components which may promote degradation, such as moisture or microbial activity.

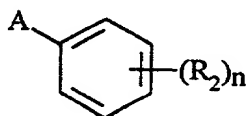
By the term «pre-emergence application» is meant an application to the soil in which the weed seeds or seedlings are present before emergence of the crop. One example of a pre-emergence application is known as «pre-plant incorporated» (PPI), where the herbicide is incorporated into the soil before planting the crop. Another is where the herbicide is applied to the soil surface after sowing the crop. By the term «foliar activity» is meant herbicidal activity produced by application to the aerial or exposed portions of the weeds which have emerged above the surface of the soil.

In general, the application rate of 4-benzoylisoxazole herbicides of formula (I) in compositions of the present invention is from 0.005 kg to 0.5 kg herbicidally active compound, preferably from 0.015 kg to 2 kg herbicidally active compound, more preferably from 0.02 kg to 0.12 kg herbicidally active compound, even more preferably from 0.05 to 0.09 kg herbicidally active compound per hectare. When sequential low doses of

isoxazole herbicide are used, as hereinbefore described, the application rates given above may be divided.

The method of the invention is generally applied to a locus pre-emergence of the weeds and crop plant. Preferably the locus is first cultivated and/or treated to remove existing weeds. For example a burn down herbicide such as glyphosate may be used.

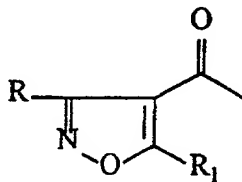
Representative herbicides whose mobility in soil is controlled by compositions of the present invention include 4-benzoylisoxazole derivatives of general formula (I):



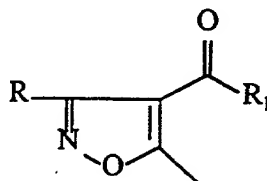
(I)

wherein:

A represents a group (A-1) or (A-2):



(A-1)



(A-2)

wherein:

R represents a hydrogen atom or a halogen atom; a straight- or branched-chain alkyl or alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; a cycloalkyl group containing from 3 to 6 carbon atoms optionally substituted by one or more groups  $R^5$ , one or more halogen atoms or a group  $-CO_2R^3$ ; or a group selected from  $-CO_2R^3$ ,  $-COR^5$ , cyano, nitro,  $-CONR^3R^4$  and  $-S(O)_kR^{13}$ ,

$R^1$  represents a straight- or branched-chain alkyl, alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; or a cycloalkyl group containing from three to six carbon atoms optionally substituted by one or more groups  $R^5$  or one or more halogen atoms;

$R^2$  represents a halogen atom; a straight- or branched-chain alkyl, alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; a straight- or branched-chain alkyl group containing up to six carbon atoms which is substituted by one or more groups  $-OR^5$ ; or a group selected from nitro, cyano,  $-CO_2R^5$ ,  $-S(O)_pR^6$ ,  $-O(CH_2)_mOR^5$ ,  $-COR^5$ ,  $-NR^{11}R^{12}$ ,  $-N(R^8)SO_2R^7$ ,  $-N(R^8)CO_2R^7$ ,  $-OR^5$ ,  $-OSO_2R^7$ ,  $-SO_2NR^3R^4$ ,  $-CONR^3R^4$ ,  $-CSNR^3R^4$ ,  $-(CR^9R^{10})_t-S(O)_qR^7$  and  $-SF_5$ ; or two groups  $R^2$ , on adjacent carbon atoms of the phenyl ring may, together with the carbon atoms to which they are attached, form a 5 to 7 membered saturated or unsaturated heterocyclic ring containing up to three ring heteroatoms selected from nitrogen, oxygen and sulfur, which ring is optionally substituted by one or more groups selected from halogen, nitro,  $-S(O)_pR^{13}$ ,  $C_{1-4}$  alkyl,  $C_{1-4}$  alkoxy,  $C_{1-4}$  haloalkyl,  $C_{1-4}$  haloalkoxy,  $=O$  (or a 5- or 6- membered cyclic acetal thereof), and  $=NO-R^3$ , it being understood that a sulphur atom, where present in the ring, may be in the form of a group  $-SO-$  or  $-SO_2-$ ;

$n$  represents an integer from one to five: when  $n$  is greater than one the groups  $R^2$  may be the same or different;

$R^3$  and  $R^4$  each independently represent a hydrogen atom, or a straight- or branched chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms;

$R^5$  represents a straight- or branched-chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; or a straight- or branched-chain alkenyl or alkynyl group containing



from two to six (preferably from three to six) carbon atoms which is optionally substituted by one or more halogen atoms;

$R^6$  and  $R^7$ , which may be the same or different, each represent  $R^5$  or phenyl optionally substituted by from one to five groups which may be the same or different selected from a halogen atom, a straight- or branched-chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms, nitro, cyano,  $-CO_2R^5$ ,  $-S(O)_pR^{13}$ ,  $-NR^{11}NR^{12}$ ,  $-OR^5$  and  $-CONR^3R^4$ ;

$R^8$ ,  $R^9$  and  $R^{10}$  each represent a hydrogen atom or  $R^6$ ;

$R^{11}$  and  $R^{12}$  each represent hydrogen or  $R^5$ ;

$R^{13}$  represents a straight- or branched-chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms;

k, p and q independently represent the values zero, one or two;

m represents one, two or three;

t represents an integer from one to four; when t is greater than one, the groups  $R^9$  and  $R^{10}$  may be the same or different;

or an agriculturally acceptable salt or metal complex thereof.

In certain cases, the groups R to  $R^{13}$  may give rise to optical and/or stereoisomerism. All such forms are embraced by the present invention.

By the term "agriculturally acceptable salts" is meant salts the cations or anions of which are known and accepted in the art for the formation of salts for agricultural or horticultural use. Preferably the salts are water soluble. Suitable acid addition salts, formed by compounds of formula (I) containing an amino group, include salts with inorganic acids, for example, hydrochlorides, sulphates, phosphates and nitrates, and salts with organic acids, for example, acetic acid. Suitable salts formed by compounds of formula (I) which are acidic, i.e. compounds containing one or more carboxy groups, with bases include alkali metal (e.g. sodium and potassium) salts, alkaline earth metal (e.g. calcium and magnesium) salts, ammonium and

amine (e.g. diethanolamine, triethanolamine, octylamine, dioctylmethylaniline and morpholine) salts.

In the description unless otherwise specified 'halogen' means a fluorine, chlorine, bromine or iodine atom.

5 Compounds of formula (I) wherein A represents (A-1) are preferred.

The phenyl ring of the compounds of formula (I) is preferably 2,4-disubstituted, 2,3-disubstituted or 2,3,4-trisubstituted.

Compounds of formula (I) in which R represents hydrogen or  $-\text{CO}_2\text{R}^3$  wherein  $\text{R}^3$  represents or a straight- or branched chain alkyl group containing up to three carbon atoms; and  $\text{R}^1$  represents cyclopropyl are preferred.

Compounds of formula (I) in which  $\text{R}^2$  represents a halogen atom; a straight- or branched chain alkyl group containing up to three carbon atoms which is optionally substituted by one or more halogen atoms;  $-\text{S}(\text{O})_p\text{R}^6$ ;  $-\text{OR}^5$  or  $-\text{CH}_2\text{S}(\text{O})_q\text{R}^7$ ; wherein  $\text{R}^5$ ,  $\text{R}^6$  and  $\text{R}^7$  each represent the same or different optionally halogenated methyl or ethyl groups are preferred.

A preferred class of compounds of formula (I) wherein A represents (A-1) are those wherein:

R is hydrogen or  $-\text{CO}_2\text{Et}$ ;

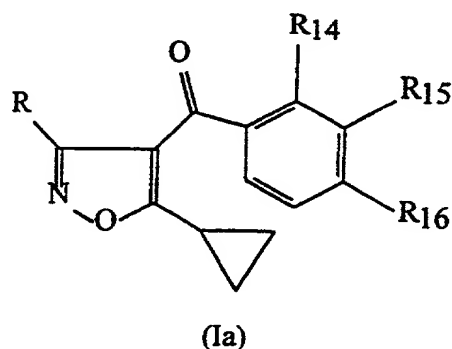
$\text{R}^1$  is cyclopropyl;

20 and two groups  $\text{R}^2$ , on adjacent carbon atoms of the phenyl ring may, together with the carbon atoms to which they are attached, combine to form a 5 or 6 membered saturated or unsaturated heterocyclic ring which is fused to the 2,3 or 3,4 positions of the benzoyl ring; wherein the heterocyclic ring contains two hetero atoms selected from sulphur and oxygen which are located at the 2 and 3, or 3 and 4 positions of the benzoyl ring; and in which the 4-substituent of the benzoyl ring is halogen or  $\text{S}(\text{O})_p\text{Me}$ , or the 2-substituent of the benzoyl ring is methyl,  $\text{S}(\text{O})_p\text{Me}$  or  $-\text{CH}_2\text{S}(\text{O})_q\text{Me}$  respectively; and optionally the heterocyclic ring may be substituted by one or more halogen atoms.

30 A more preferred class of compounds of formula (I) are those wherein A represents (A-1); R is hydrogen or  $-\text{CO}_2\text{Et}$ ;  $\text{R}^1$  is cyclopropyl;  $\text{R}^2$  is a

halogen atom or a group selected from  $-\text{CF}_3$ , Me, Et,  $-\text{S}(\text{O})_p\text{Me}$ ,  $-\text{CH}_2\text{S}(\text{O})_q\text{Me}$  and optionally halogenated methoxy or ethoxy; and n is two or three.

A yet more preferred class of compounds of formula (I) are those having the formula (Ia):



wherein:

R is hydrogen or  $-\text{CO}_2\text{Et}$ ;

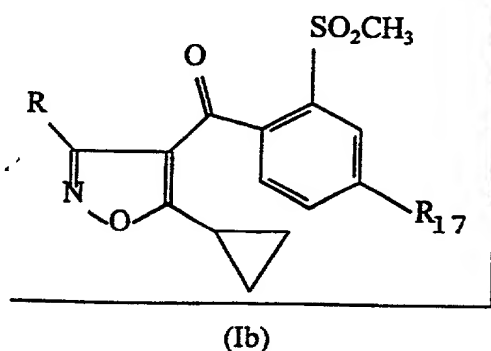
$\text{R}^{14}$  is selected from  $-\text{S}(\text{O})_p\text{Me}$ , Me, Et, a chlorine, bromine or fluorine atom, methoxy, ethoxy and  $-\text{CH}_2\text{S}(\text{O})_q\text{Me}$ ;

$\text{R}^{15}$  is selected from a hydrogen atom, a chlorine, bromine or fluorine atom, methoxy, ethoxy and  $-\text{S}(\text{O})_p\text{Me}$ ; and

$\text{R}^{16}$  is selected from a hydrogen atom, a chlorine, bromine or fluorine atom, methoxy and  $\text{CF}_3$ ;

and wherein at least one of  $\text{R}^{15}$  and  $\text{R}^{16}$  is other than hydrogen.

An especially preferred class of compounds of formula (I) have the formula (Ib):



wherein R<sup>17</sup> is chlorine, bromine or trifluoromethyl; and  
R is hydrogen or -CO<sub>2</sub>Et.

The following compounds of formula (I) are among the most preferred  
for use in the present invention:

5        5-cyclopropyl-4-[2-chloro-3-ethoxy-4-(ethylsulphonyl)benzoyl]isoxazole;  
      4-(4-chloro-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;  
      5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole;  
      4-(4-bromo-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;  
      5-cyclopropyl-4-[4-fluoro-3-methoxy-2-(methylsulphonyl)benzoyl]isoxazole;  
10       4-(4-bromo-2-methylsulphonylmethylbenzoyl)-5-cyclopropylisoxazole;  
      ethyl 5-cyclopropyl-4-(2-methylsulphonyl-4-  
trifluoromethylbenzoyl)isoxazole-3-carboxylate;  
      5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)-3-methylthio-  
isoxazole.

15       The most preferred compound is 5-cyclopropyl-4-(2-methylsulphonyl-  
4-trifluoromethylbenzoyl)isoxazole (isoxaflutole).

Procedures for preparing isoxazoles of formula (I) are as described in  
European Patent Publication Nos. 0418175, 0487357, 0527036 and 0560482.

20       The method of the invention can be used on genetically modified crops.  
      By genetically modified crop is understood those crops which have  
been made tolerant towards herbicides by conventional sowing and  
cultivation methods or genetic engineering methods.

25       According to a further feature of the present invention, there are  
provided compositions suitable for herbicidal use in the method of the  
invention comprising one or more of the 4-benzoylisoxazoles of formula (I)  
or an agriculturally acceptable salt or metal complex thereof (which may be  
encapsulated as hereinbefore described), in association with, and preferably  
homogeneously dispersed in, one or more compatible agriculturally-  
30       acceptable diluents or carrier and/or surface active agents [i.e. diluents or  
carriers and/or surface active agents of the type generally accepted in the art  
as being suitable for use on herbicidal compositions and which are

compatible with compounds of formula (I)]. The term «homogeneously dispersed» is used to include compositions in which the compounds of formula (I) are dissolved in other components. The term «herbicidal compositions» is used in a broad sense to include not only compositions which are ready for use as herbicides but also concentrates which must be diluted before use. Preferably, the compositions contain from 0.05 to 90% by weight of one or more compounds of formula (I).

The herbicidal compositions may contain both a diluent or carrier and surface-active (e.g. wetting, dispersing, or emulsifying) agent. Surface-active agents which may be present in herbicidal compositions of the present invention may be of the ionic or non-ionic types, for example sulphoricinoleates, quaternary ammonium derivatives, products based on condensates of ethylene oxide with alkyl and polyaryl phenols, e.g. nonyl- or octyl-phenols, tristyryl phenols, condensates of ethylene oxide with alcohols, or carboxylic acid esters of anyhydrosorbitols which have been rendered soluble by etherification of the free hydroxy groups by condensation with ethylene oxide, alkali and alkaline earth metal salts of sulphuric acid esters and sulphonic acids such as dinonyl- and dioctyl-sodium sulphonosuccinates and alkali and alkaline earth metal salts of high molecular weight sulphonic acid derivatives such as sodium and calcium lignosulphonates and sodium and calcium alkylbenzene sulphonates.

Suitably, the herbicidal compositions according to the present invention may comprise up to 10% by weight, e.g. from 0.05% to 10% by weight, of surface-active agent but, if desired, herbicidal compositions according to the present invention may comprise higher portions of surface-active agent, for example up to 15% by weight in liquid emulsifiable suspension concentrates and up to 25% by weight in liquid water soluble concentrates.

Examples of suitable solid diluents or carriers are aluminium silicate, microfine silicon dioxide, talc, chalk, calcined magnesia, kieselguhr, tricalcium phosphate, powdered cork, adsorbent carbon black and clays such as kaolin, attapulgite, diatomaceous earth, mica, alumina oxide, titanium oxide and bentonite. The solid compositions (which may take the form of

dusts, granules or wettable powders) are preferably prepared by grinding the compounds of formula (I) with solid diluents or by impregnating the solid diluents or carriers with solutions of the compounds of formula (I) in volatile solvents, evaporating the solvents and if necessary, grinding the products so as to obtain powders. Granular formulations may be prepared by absorbing the compounds of formula (I) dissolved in suitable solvents, (which may, if desired, be volatile) onto the solid diluents or carriers in granular form and, if desired, evaporating the solvents, or by granulating compositions in powder form obtained as described above. Solid herbicidal compositions, particularly wettable powders and granules, may contain wetting or dispersing agents (for example of the types described above), which may also, when solid, serve as diluents or carriers.

Liquid compositions according to the invention may take the form of aqueous, organic or aqueous-organic solutions, suspensions and emulsions which may incorporate a surface-active agent. Suitable liquid diluents for incorporation in the liquid compositions include water, glycols, glycol ethers, tetrahydrofurfuryl alcohol, acetophenone, cyclohexanone, isophorone, alkyl pyrrolidones, butylolactone, chlorinated toluene, xylene, mineral, animal and vegetable oils, esterified vegetable oils and light aromatic and naphthenic fractions of petroleum (and mixtures of these diluents). Surface-active agents, which may be present in the liquid compositions, may be ionic or non-ionic (for example of the types described above) and may, when liquid, also serve as diluents or carriers.

Powders, dispersible granules and liquid compositions in the form of concentrates may be diluted with water or other suitable diluents, for example mineral or vegetable oils, particularly in the case of liquid concentrates in which the diluent or carrier is an oil, to give compositions ready for use.

When desired, liquid compositions of the compounds of formula (I) may be used in the form of self-emulsifying concentrates containing the active substances dissolved in the emulsifying agents or in solvents containing emulsifying agents compatible with the active substances, the

simple addition of such concentrates to water producing compositions ready for use.

Liquid concentrates in which the diluent or carrier is an oil may be used without further dilution using the electrostatic spray technique.

5           Herbicidal compositions according to the present invention may also contain, if desired, conventional adjuvants such as adhesives, protective colloids, thickeners, penetrating agents, spreading agents, stabilisers, buffers, sequestering agents, anti-caking agents, colouring agents and corrosion inhibitors. These adjuvants may also serve as carriers or diluents.

10           Unless otherwise specified, the following percentages are by weight. Preferred herbicidal compositions according to the present invention are encapsulations containing water dispersible granules which comprise from 1 to 90%, e.g. 25 to 75% of one or more compounds of formula (I), from 1 to 15%, e.g. 2 to 10%, of surface-active agent and from 5 to 95%, e.g. 20 to 60%, of solid diluent, e.g. clay, granulated with the addition of water to form a paste and then dried;

15           aqueous suspension concentrates which comprise from 5 to 70% of one or more compounds of formula (I), from 2 to 10% of surface-active agent, from 0.1 to 5% of thickener and from 15 to 87.9% of water;

20           wetable powders which comprise from 5 to 90% of one or more compounds of formula (I), from 2 to 10% of surface-active agent and from 8 to 88% of solid diluent or carrier;

25           water soluble or water dispersible powders which comprise from 5 to 90% of one or more compounds of formula (I), from 2 to 40% of sodium carbonate and from 0 to 88% of solid diluent;

30           liquid water soluble concentrates which comprise from 5 to 50%, e.g. 10 to 30% of one or more compounds of formula (I), from 0 to 25% of surface-active agent and from 10 to 90%, e.g. 45 to 85%, of water miscible solvent, e.g. triethylene glycol, or a mixture of water-miscible solvent and water;

            liquid emulsifiable suspension concentrates which comprise from 5 to 70% of one or more compounds of formula (I), from 5 to 15% of surface-

active agent, from 0.1 to 5% of thickener and from 10 to 84% of organic solvent, e.g. mineral oil; and

emulsifiable concentrates which comprise 0.05 to 90%, and preferably from 1 to 60% of one or more compounds of formula (I), from 0.01 to 10%, and preferably from 39 to 98.99%, of organic solvent.

The water dispersible granules comprising isoxazoles of formula (I) whose apparent density was 0.25 – 0.75, have a particle size of generally 10-2000  $\mu\text{m}$ , preferably 300-1500  $\mu\text{m}$ .

Herbicidal compositions according to the present invention may also comprise the compounds of formula (I) in association with, and preferably homogeneously dispersed in, one or more other pesticidally active compounds and, if desired, one or more compatible pesticidally diluents or carriers, surface-active agents and conventional adjuvants as hereinbefore described.

Examples of other pesticidally active compounds which may be included in, or used in conjunction with, the herbicidal compositions of the present invention include herbicides, for example to increase the range of weed species controlled for example acetochlor,

alachlor [2-chloro-2,6'-diethyl-N-(methoxy-methyl)-acetanilide], atrazine [2-chloro-4-ethylamino-6-isopropylamino-1,3,5-triazine], bromoxynil [3,5-dibromo-4-hydroxybenzonitrile], chlortoluron [N'-(3-chloro-4-methylphenyl)-N,N-dimethylurea], cyanazine [2-chloro-4-(1-cyano-1-methylethylamino)-6-ethylamino-1,3,5-triazine], 2,4-D [2,4-dichlorophenoxy-acetic acid], dicamba [3,6-dichloro-2-methoxybenzoic acid], difenzoquat [1,2-diethyl-3,5-diphenyl-pyrazolium salts], dimethanamid, flampropmethyl [methyl N-2-(N-benzoyl-3-chloro-4-fluoroanilino)-propionate], flufenacet, fluometron [N'-(3-trifluoromethylphenyl)-N,N-dimethylurea], glyphosate, glufosinate, isoproturon [N'-(4-isopropylphenyl)-N,N-dimethylurea], metolachlor, metribuzin, insecticides, e.g. synthetic pyrethroid, e.g. permethrin and cypermethrin, fipronil and fungicides, e.g. carbamates, e.g. methyl N-(1-butyl-carbamoyl-



benzimidazol-2-yl)carbamate, and triazoles e.g. 1-(4-chloro-phenoxy)-3,3-dimethyl-1-(1,2,4-triazol-1-yl)-butan-2-one.

Pesticidally active compounds and other biologically active materials which may be included in, or used in conjunction with, the herbicidal compositions of the present invention, for example those hereinbefore mentioned, and which are acids, may, if desired, be utilised in the form of conventional derivatives, for example alkali metal and amine salts and esters.

The following Examples illustrate herbicidal compositions which may be used in the present invention. The Active Ingredient listed in the following examples refers to a compound of general formula (I).

**Example C1:**

An emulsifiable concentrate is formed from:

Active ingredient	20% w/v
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N-Methylpyrrolidinone (NMP)	25% w/v
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Calcium dodecylbenzenesulphonate (CaDDBS)	4% w/v
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Nonylphenol ethylene oxide propylene oxide

Condensate (NPEOPO)	4% w/v
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Aromatic solvent	to 100 volumes
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by stirring NMP, active ingredient (Compound 1), CaDDBS, NPEOPO and Aromatic solvent until a clear solution is formed, and adjusting to volume with Aromatic solvent.

**Example C2**

A wettable powder is formed from:

Active Ingredient	50% w/w
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Sodium dodecylbenzenesulphonate	3% w/w
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Sodium methyl oleoyl taurate	5% w/w
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Sodium polycarboxylate	1% w/w
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Microfine silicon dioxide	3% w/w
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China clay	38% w/w
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by blending the above ingredients together and grinding the mixture in an air jet mill.

### **Example C3**

5 A suspension concentrate is formed from:

Active Ingredient	50% w/v
Antifreeze (Propylene glycol)	5% w/v
Ethoxylated tristyrylphenol phosphate	0.5% w/v
Nonyl phenol 9 mole ethoxylate	0.05% w/v
10 Sodium polycarboxylate	0.02% w/v
Attaclay	1.5% w/v
Antifoam	0.003% w/v
Water	to 100 volumes

by stirring the above ingredients together and milling in a bead mill.

### **Example C4**

15 A water dispersible granule is formed from:

Active Ingredient	50% w/w
Sodium dodecylbenzenesulphonate	3% w/w
20 Sodium methyl oleoyl taurate	5% w/w
Sodium polycarboxylate	1% w/w
Binder (Sodium lignosulphonate)	8% w/w
china clay	30% w/w
Microfine silicon dioxide	3% w/w

25 by blending the above ingredients together, grinding the mixture in an air jet mill and granulating by addition of water in a suitable granulation plant (e.g. Fluid bed drier) and drying. Optionally the active ingredient may be ground either on its own or admixed with some or all of the other ingredients.

30 The following non-limiting Example illustrates the invention.

### **EXAMPLE 1**

Glasshouse experiment showing unexpected enhancement of biological activity of weeds following application of compound (isoxaflutole).

7 x 7 cm<sup>2</sup> pots were filled with a non-sterile loam soil. Weed seeds (*Amaranthus retroflexus*, *Echinochloa crus galli* and *Setaria viridis*) were placed in three separate shallow wells with a maize seed (Pioneer 3394) inserted to a depth of 4 cm in between in each pot and the seeds lightly covered with soil.

A 1 ml solution of technical isoxaflutole, suitably diluted to give a dose equivalent to 6.25, 12.5, 25, 50 and 100 g/ha in acetonitrile, was pipetted evenly on the soil surface of pots (replicated 10 times), according to the following regime.

On day 1, a set of 5 pots were treated with 5 dose rates (6.25 – 100 g/ha) and set aside in the glasshouse. At the same time, on day 1 a second set of 4 pots were treated with 4 dose rates (treatment A: 6.25-50 g/ha) along with a third set of 3 pots treated with 3 dose rates (treatment B: 6.25-25 g/ha). On day 2 each of the second and third pots were treated again the pots receiving identical treatments A and B and second set of pots set aside in the glasshouse. On day 3 each of the third set of pots were treated again, the pots receiving identical treatment B. On day 4 each of the third set of pots were treated, the pots receiving identical treatment B and the pots set aside in the glasshouse.

The pots were maintained in a glasshouse, with overhead watering (3 x daily) and supplementary lighting. Visual assessment of % reduction, compared to untreated control plants was recorded 14 days after treatment. The results (average of 10 replicates) are shown on Table 1, where the dosing regime refers to the number of daily applications followed by the dose of compound on each day.

TABLE 1

% Damage 14 DAT: Comparison of Single vs. Multiple Application

% Damage				
Dosing Regime	Amare	Echeg	Setvi	Maize (P 3394)
2 x 6.25	34	18	18	0
1 x 12.5	0	0	0	0
4 x 6.25	80	97	75	0
2 x 12.5	46	68	42	0
1 x 25	8	32	10	0
4 x 12.5	79	100	91	6
2 x 25	42	70	44	0
1 x 50	50	84	24	2
4 x 25	90	100	77	6
2 x 50	78	96	66	10
1 x 100	79	94	59	8

As can be seen in Table 1, the efficacy of the herbicide on key weed species is improved by 2 applications separated by 1 day and surprisingly even more enhanced by 4 applications separated by 3 days, compared to a single application of the same total dose of compound. Repeat applications did not appear to have any impact on maize phytotoxicity.

## EXAMPLE 2

A 30% aqueous solution of Gerol was prepared and 67 g added to 80 g of a suspension of isoxaflutole (250 g/L) in water containing a dispersant (sodium polynaphthalene sulphate to maintain the suspension) with stirring. Powdered calcium carbonate (1 g) was added and the resultant mixture sonicated with ultrasound before addition of polysiloxane oil (600 ml) to give an emulsion. Acetic acid (3 ml) was added and stirring continued for 2 hours. The stirring was stopped and the upper layer decanted from the precipitate. This solid was filtered off, washed with water and dried to give

microparticles of encapsulated isoxaflutole. The encapsulated material may be formulated using methods hereinbefore described.

- 5 Gerol: Diethyleneglycol-ethyleneglycol-isophthalic acid-sodium 5-sulfoisophthalate-terephthalic acid-triethylene glycol copolymer.

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**EXAMPLE 3**

5           Maize is sown and then grown up in area where weeds and weed seeds are present. The weeds are a selection from: Alopecurus myosuroides, Avena fatua, Digitaria sanguinalis, Echinochloa crus-galli, Eleusine indica, Lolium multiflorum, Setaria viridis, Sorghum halepense, Cyperus esculentus, Cyperus iria, Cyperus rotundus, Eleocharis acicularis, Abutilon theophrasti,  
10           Amaranthus retroflexus, Bidens pilosa, Chenopodium album, Galium aparine, Ipomoea purpurea, Lamium purpureum, Matricaria inodora, Sesbania exalta, Sinapis arvensis, Solanum nigrum, Stellaria media, Veronica hederifolia, Veronica Persia, Viola arvensis and Xanthium strumarium,

          After one week sowing the maize, isoxazole is sprayed as an  
15           encapsulated formulation as hereinbefore described at a rate of 105 g/ha of herbicidally active compound, the weight ratio of isoxazole: copolyester being 1:10. The amounts of isoxazole of formula (I) and DKN were measured after 4 days in a 5 cm deep soil core around the seed or seedling. The weight ratio of isoxazole: DKN was found to be 1.

20           The activity of the isoxazole on the crop and weeds was observed after 3 weeks, and found to be equal to 2% and 95% respectively.

          A similar application in similar soil conditions without the copolyester provided a weight ratio of isoxazole: DKN of 0.1 and herbicidal activity on both crop and weeds was 15% and 95% respectively.

25

**EXAMPLE 4**

          Maize is sown in an area where weeds and weed seeds of the species Setaria viridis are present. Isoxazole is sprayed on the soil surface as an  
30           encapsulated formulation at a rate of 105 g herbicidally active compound per hectare. The activity of the isoxazole on maize and Setaria viridis was observed 6, 11, 14 and 17 days after treatment (DAT).

**Activity of 4-benzoylisoxazole on *Setaria viridis***

Application rate g/ha	Formulation	DAT			
		6	11	14	17
105	Encapsulated	0	40	70	90
	WG	10	50	75	100

**Activity of 4-benzoylisoxazole on Maize**

Application rate g/ha	Formulation	DAT			
		6	11	14	17
105	Encapsulated	0	0	3	8
	WG	0	5	7	10

5           Activity on *Setaria viridis* by the encapsulated formulation was equal to that of the WG formulation. The encapsulated formulation decreased phytotoxicity on maize by 20-30% compared to the WG formulation.

**EXAMPLE 5**

10

15

Maize is sown and grown up in an area where weeds and weed seedlings are present. The weeds are *Amaranthus retroflexus*, *Echinochloa crus-galli* and *Setaria viridis*. Solutions of isoxazole in acetonitrile were applied directly to the soil surface at dose rates of 100, 50 and 25 g/ha to plots A, B and C respectively. 1 DAT dose rates of 50 and 25 g/ha were applied to plots B and C respectively, 2 and 3 DAT a dose rate of 25 g/ha was applied to plot C.

Activity of the isoxazole on maize and weed species was observed 14 DAT.

Dosing regime	AMARE	ECHCG	SETVI	Maize
4 x 25 g/ha	90	100	77	6
2 x 50 g/ha	78	96	66	10
1 x 100 g/ha	79	94	59	8

- 5 Maintaining the isoxazole in the surface of the soil within the area containing weed seeds and seedlings, by sequential application of low dose rates of isoxazole, provides improved activity on weeds.

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## CLAIMS

1                   .           A method for controlling the growth of weeds at a  
locus in a solid growing medium which comprises treating the locus with a  
5                   composition comprising an isoxazole herbicide to provide progressive or  
sequential delivery or release of isoxazole herbicide into the surface layer of  
the medium.

2                   .           A method according to claim 1 in which the  
growing medium is soil.

10                  3                   .           A method according to claim 1 or 2 in which the  
locus is a crop-growing locus.

4                   .           A method according to claim 1, 2 or 3 in which the  
surface layer of the medium is from the surface to a depth of 10 cm.

15                  5                   .           A method according to any one of the preceding  
claims which comprises applying to the locus sequential low doses of  
isoxazole herbicide.

20                  6                   .           A method according to any one of claims 1 to 4  
which comprises treating the locus with a delayed release composition  
comprising the isoxazole herbicide.

25                  7                   .           A method according to claim 6 in which the  
delayed release composition comprises an encapsulated composition.

8                   .           A method according to claim 6 or 7 in which an  
encapsulated isoxazole is used, comprising an isoxazole derivative  
encapsulated with a solid film comprising an inert material itself having no  
substantial herbicidal activity.

30                  9                   .           A method according to claim 8 in which granules of  
an isoxazole derivate from 0.1 to 50  $\mu\text{m}$  in size are used.

10. A method according to any one of the preceding claims in which the isoxazole derivative is of general formula I as hereinbefore defined.

11. A delayed release composition comprising an isoxazole  
herbicide.

12. A method according to claim 1 substantially as  
hereinbefore described in Example 1.

10           13. A delayed release composition according to claim 9 substantially  
as hereinbefore described.

[illegible]



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/EP00/01102 <b>(22) International Filing Date:</b> 1 February 2000 (01.02.00)  <b>(30) Priority Data:</b> 9902232.9                      1 February 1999 (01.02.99)                      GB 9908313.1                      12 April 1999 (12.04.99)                      GB  <b>(71) Applicant (for all designated States except US):</b> AVENTIS AGRICULTURE LIMITED [GB/GB]; Fyfield Road, Ongar, Essex CM5 0HW (GB).  <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> ROBERTS, David, Alan [GB/GB]; Aventis Agriculture Ltd, Research Station - Fyfield Road, Ongar, Essex CM5 0HW (GB). ZERROUK, Robert [FR/FR]; La Combe de Berg, F-43220 Dunières (FR). COLEGATE, Rachel [GB/GB]; Aventis Agriculture Ltd, Fyfield Road, Ongar, Essex CM5 0HW (GB).  <b>(74) Agent:</b> BRACHOTTE, Charles; Rhône-Poulenc Agro, Département Propriété Industrielle, Boîte postale 9163, F-69263 Lyon Cedex 09 (FR).		<b>(81) Designated States:</b> AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i> <i>Before the expiration of the time limit for amending the</i> <i>claims and to be republished in the event of the receipt of</i> <i>amendments.</i>
<b>(54) Title:</b> WEED CONTROL		
<b>(57) Abstract</b>  The invention provides a method for controlling the growth of weeds at a locus in a solid growing medium which comprises treating the locus with a composition comprising an isoxazole herbicide to provide progressive or sequential delivery or release of isoxazole herbicide into the surface layer of the medium.		

# COMBINED DECLARATION AND POWER OF ATTORNEY

Attorney Docket No.

PH 99012G1

5500\*99

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled "Weed Control"

the specification of which

(check one) ☐ is attached hereto.

☒ was filed on July 30, 2001 as

Application Serial No. 09/890,384 and

was amended on July 30, 2001  
(if applicable)

was amended through \_\_\_\_\_  
(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed:

## Prior Foreign Application(s)

## Priority Claimed

<u>PCT/EP00/01102</u> (Number)	<u>International</u> (Country)	<u>1 February 2000</u> (Day/Month/Year Filed)	<input checked="" type="checkbox"/> <input type="checkbox"/> Yes No
<u>9908313.1</u> (Number)	<u>Great Britain</u> (Country)	<u>12 April 1999</u> (Day/Month/Year Filed)	<input checked="" type="checkbox"/> <input type="checkbox"/> Yes No
<u>9902232.9</u> (Number)	<u>Great Britain</u> (Country)	<u>1 February 1999</u> (Day/Month/Year Filed)	<input checked="" type="checkbox"/> <input type="checkbox"/> Yes No

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status)
_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status) (patented, pending, abandoned)

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below.

\_\_\_\_\_  
(Application No.)      \_\_\_\_\_  
(filing date)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.



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In the matter of the above-identified application, please recognize the attorneys associated with **CUSTOMER NUMBER 23416** all of CONNOLLY BOVE LODGE & HUTZ LLP, as attorneys with full power of substitution to prosecute this application and conduct all business in the Patent and Trademark Office connected therewith.

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